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SEARCH FOR AND STUDY OF NOVEL SUPERCONDUCTOR WITH HIGHER TC AND JC

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UNIVERSITY OF HOUSTON SYSTEM

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14. ABSTRACT
The AFOSR support has enabled us to discover new superconductors with higher Tc, for science and potential new applications, or with low Tc but with novel structure-types for new discoveries; to realize new superconducting mechanism for higher Tc; to unravel new physics important to high Tc and Jc; to discover new physics and phenomena in non-superconducting but related compounds for HTS development and devices; to develop single-crystal growth capability and new techniques; to evaluate immediately new discoveries; and to compete effectively against foreign HTS groups. A record Tc up to 49K was discovered in the rare-earth doped CaFe2As2 system, highest in the Fe-based 122 family; the unusually high Tc is attributed to the possible interfacial enhancement mechanism, opening up a new avenue to higher Tc. Interface effect in the artificially grown FeSe/STO system was carefully studied. More than ten new superconductors were discovered, and provide new directions and new synthetic routes to search for higher Tc. New physics and phenomena were discovered in both superconducting and non-superconducting compounds including cuprates, other Fe-based superconductors, multiferroics, topological insulators, and thermoelectric and thermal-conducting materials.

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AFOSR Final Report: 9/15/2009 - 9/14/2015
Title: Search for and Study of Novel Superconductor with Higher T_c and J_c
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Accomplishments

- Synthesized and characterized large single crystals of rare-earth doped (Ca,RE)122 system with RE = , La, Ce, Pr and Nd, with onset-T_c of 42-49 K, higher than any compounds *R*-Ca-Fe-As previously known, and representing as the highest T_c in the Fe-based 122 family.
- Demonstrated the non-bulk nature, two superconducting transitions, and unusually high magnetic anisotropy in the (Ca,RE)122 system. The high onset-T_c is also found to be RE-independent and dopant concentration independent.
- Carried out detailed chemical composition, magnetization, transport, low temperature annealing effects, Raman, ARPES, and STM studies for the (Ca,RE)122 system, excluded the possibilities of the 49K SC transition being caused by an optimally doped 122, an oxygen-deficient 1111 phase due to contamination by oxygen, or an unknown residual-strain-induced precipitate resulting from the collapsed tetragonal phase transition, and attributed the cause for the unusually high T_c to the possible long-sought-after interfacial enhancement mechanism of naturally grown FeAs layers with different defect structures.
- Performed detailed dc, ac, sheet resistance and aging effect measurements on the 1-4 UC FeSe/STO thin films synthesized by Xue's group in Tsinghua, using a novel technique developed by us and observed: a Meissner state below ~ 20 K; a mesoscopic state with superconducting patches between ~ 20 and 45 K; and a yet-to-be-determined state between ~ 45 and 80 K with a logarithmic frequency dispersion.
- Designed a Fully Integrated MBE (Growth) and Characterization System (AFM/STM, ARPES, Magneto-resistance) to further investigate the interface superconductivity mechanism for different materials. The system will have the following specifications: (I) MBE thin film growth with (II) a load lock chamber, (III) a sample preparation chamber with the capability to accommodate an ARPES characterization system at a later stage, (IV) an STM/AFM chamber, and (V) a magneto-resistance chamber to be added later, with the help of world-wide MBE experts (Q. K. Xue in China and I. Bozovic at BNL) and in-house expert (A. Bensaoula). The system is expected to be delivered in September 2016 and we are ready to fabricate high quality thin films.
- Discovered the new superconductor $(Ba,Na)Ti_2Sb_2O$ with T_c up to 5.5 K, displaying anti-perovskite Ti_2O layer structure (as compared to the CuO_2 planes in HTS).
- Discovered the new superconductor Zr_5Sb_3 with a T_c of 2.3 K, the first superconductor discovered by us in a compound system of Mn_5Si_3-structure type that consists of a large number of compounds.
- Discovered a series of Pt-related new superconductors with distinct structure features: $SrPt_3P$ (T_c =8.6K) with anti-perovskite structure, $SrPt_6P_2$ (T_c= 0.6K) with anti-trigonal prism structure, and $SrPt_{10}P_4$ (T_c=1.5K) with new type of structure.
- Systematically grew and studied a series of Pd-based new superconductors: layered compound β-$PdBi_2$, hole-doped $PdBi_{2-x}Pb_x$, and electron-doped Na_xPdBi_2 single crystals with maximum T_c at 5.5K.

- Induced bulk superconductivity up to 4 K in $ZrTe_3$ single crystals without chemical doping or physical pressure by processing induced-defects, and provided a new route to induce superconductivity in other materials such as multiferroics.
- Discovered precursor-dependent superconducting/nonsuperconducting properties in the $Rb_xFe_2Se_2$ single crystals with T_c up to 32-K.
- Carried out systematic pressure study up to 1.8GPa on all of the compounds mentioned above. Besides the above compounds, we also studied the pressure dependence in both superconducting compounds such as $FeSe_{0.5}Te_{0.5}$ thin films and Fe pnictide 122s, and non-superconducting compounds such as $SrCu_4As_2$, $Sr_{1-x}K_xCu_4As_2$, and $R$$Fe_2Si_2$.
- Synthesized the topological insulator Bi_2Se_3, and 3.8-K $Cu_xBi_2Se_3$ “topological superconductor” single crystals, carried out nanoscale Andreev reflection spectroscopy study on $Cu_xBi_2Se_3$ and called for a reexamination of the reported signature of topological superconductivity in $Cu_xBi_2Se_3$.
- Discovered a drastic superconducting-semiconducting transition in YBCO at ~ 30 GPa.
- Demonstrated that the pressure induced SC state in Ca122 is bulk through specific heat measurement under press, in contrast to previous suggestion.
- Continued to carry out systematically magnetic, resistivity, specific heat, Hall effect, high pressure, annealing effect to study both the normal and superconducting properties of different types of Fe-based superconductors.
- Attempted to look for superconductivity in the other 122 system without pnictides such as RFe_2Si_2 and $R(Fe_{1-x}M_x)_2Si_2$(x = 0-1) systems (R = La, Y and Lu, M = Ni, Mn and Cu). No superconductivity is observed, although some interesting long-range magnetic order is observed which is systemically changing with different rare earth elements and different transition metal doping.
- Attempted to look for superconductivity in Cu-based pnictide Cu-based compounds: 111 family- CaCuP, CaCuAs, SrCuP, SrCuAs, BaCuP, BaCuAs, EuCuP, EuCuAs; 122 family- $CaCu_{2-x}As_2$, $SrCu_{2-x}As_2$, $BaCu_{2-x}As_2$, $EuCu_{2-x}As_2$, $EuCu_2Sb_2$; and 142 family- KCu_4As_2, $RbCu_4As_2$, $CaCu_4As_2$, $SrCu_4As_2$, $BaCu_4As_2$, $EuCu_4As_2$, KCu_4As_2, $NaCu_4Sb_2$, and KCu_4Sb_2, in polycrystalline and single crystalline form, and some selected doping studies as well.
- Discovered two completely new phase of $BaCu_2As_2$ deriving from the typical $ThCr_2Si_2$ type with very sensitive temperature-dependent formation mechanism.
- Extended our research field to other AFOSR mission related system such as super-thermal-conducting materials, thermoelectric materials with higher ZT and better performance, topological insulators and superconductors, and 2D transition metal dichalcogenides.
- Provided experimental evidence of a temperature-induced orbital-selective Mott transition (OSMT) in the $A_xFe_{2-y}Se_2$ (A=K, Rb), in collaboration with Z. X. Shen’s group.
- Observed broken time reversal and parity symmetries of the superconducting state of LiFeAs through magnetic torque and magnetization measurements at high fields, in collaboration with Luis Balicas of NHMFL.
- Studied quasiparticle relaxation utilizing ultrafast optical spectroscopy in stoichiometric LiFeAs crystals and (K, Ba)Fe_2As_2 crystals in collaboration with M. K. Wu’s group at Academia Sinica.
- Calculated the Superfluid Density in the $s_{\pm}$-Wave State of Clean Iron-Based Superconductors and showed that the Uemura relation also holds for the Fe-based superconductors (Ting’s group).

- Theoretically investigated the Interaction-Induced Localization of Fermionic Mobile Impurities in a Larkin-Ovchinnikov Superfluid in a two-dimensional optical lattice and showed that the impurity atoms can localize and form pairs when the interaction between the impurity atoms and the LO superfluid is strong (Ting's group).
- Proposed the detection of time-reversal symmetry in topological surface states (Ting's group).
- Studied the 122, 111, and 11 compounds with emphasis on their electronic structures and atomic defects by our home built STM; also started to study the 111 and 11 compounds and observed unusual features that challenges existing theoretical models (Pan's group).
- Discovered the possibility of electrically charging multiferroic domain walls by applying a magnetic field to $Mn_{0.95}Co_{0.05}WO_4$.
- Found and explained a spontaneous polarization reversal in the multiferroic $Mn_{0.85}Co_{0.15}WO_4$ when cooled in external magnetic field; this property is most likely related to the formation of domains of different multiferroic phases and indicates a strong coupling across the domain walls.
- Investigated the details of the helical magnetic order of $Mn_{1-x}Co_xWO_4$ through magnetic, dielectric, and elastic neutron scattering experiments and acquired a fundamental understanding of the complex magnetic orders and the relation to ferroelectricity, have revealed the complete phase diagram showing multiple polarization flops upon Co doping.
- Determined the most fundamental magnetic exchange coupling constants of $MnWO_4$ through inelastic neutron scattering experiments and found that the interactions are long-ranged, extending to far distant neighbors.
- Studied the effect of nickel doping on the ferroelectric properties of $MnWO_4$ and found an unusual anti-correlation between the easy axis of the Ni spin and the orientation of the spin cycloid in the multiferroic phase.
- Grew large single crystals of several pure and doped multiferroics and demonstrated that the nature of dopants, including Fe, Co, Ni, Cu, Zn, can change the characteristics of the phase diagram in $MnWO_4$.
- Observed a field-induced change of the electronic band gap in the multiferroic $Ni_3V_2O_8$ single crystals grown via floating zone optical furnace.
- Studied the pressure effect on the multiferroic properties of $GdMn_2O_5$ and found a pressure-induced splitting of the ferroelectric phase into two phases, indicating a decoupling of the rare earth and Mn spin systems due to the compression of the c-axis.
- Systematically studied the role of the rare earth moment in the magnetoelectric system $RAl_3(BO_3)_4$ for R=Tb, Ho, Er, Tm and solid solutions of different compounds.
- Discovered a giant magnetoelectric effect in $HoAl_3(BO_3)_4$ setting a record for the value of the field-induced polarization at this time.
- Discovered and studied the antiferromagnetic phase and the field-induced spin-flop transition in the polar compound $LiCrP_2O_7$ (isostructural to the magnetoelectric $LiFeP_2O_7$); found that the magnetic order in $LiCrP_2O_7$ is very different from $LiFeP_2O_7$, which explains the missing magnetoelectric property in the former compound.
- Discovered an unusual magnetic order and strong magnetoelectric and magnetoelastic interactions in the polar compound $LiFeP_2O_7$, and developed a comprehensive Landau theory describing the magnetic order and the magnetoelectric effect.
- Discovered a structural and magnetic transition in the new compound $Nb_2O_2F_3$ with extremely slow transformation kinetics. Pairing of Nb-Nb dimers is observed with electrons forming spin singlet states and quenching the magnetic moment.

- Discovered and studied the magnetic order and a temperature-induced spin rotation in the transition metal-organic compound $CH(NH_2)_2Mn(HCOO)_3$; CN_2 molecules in this structure have a dipolar momentum and the question whether or not the dipoles order and give rise to a ferro-(antiferro-) electric state and multiferroic properties is of interest; first dielectric data show evidence for a dielectric transition just below room temperature.
- Studied the properties of low-dimensional, frustrated magnetic vanadate systems with super-super exchange interactions, $AAg_2M[VO_4]_2$ (A=Ba, Sr and M=Co, Ni, Cu), and discovered ferromagnetic as well as antiferromagnetic orders and novel magnetic properties.
- Determined the quantum criticality in doped Sr and Ba122.
- Discovered the existence of quantum (Shubnikov-de Haas) oscillations in metallic $Bi_2(Te,Se)_3$ arising from topological surface as well as trivial bulk states in different ranges of the applied magnetic field. This solves the conundrum why the surface oscillations have been so clearly observed by us in metallic $Bi_2(Te,Se)_3$.
- Detected weak antilocalization effects in Bi_2Te_3 topological insulators. We could show that the charge carrier density, but not their nature (electrons or holes), determine whether or not the antilocalization effects arise from bulk or topological surface states.
- Found quantum oscillations from topological surface states in the new topological insulator Sb_2Te_2Se. Here the quantum oscillations are very pronounced and no bulk oscillations could be detected.

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J. Shulman, Ph.D., Physics, December 2009, Dissertation: "A general mechanism for negative capacitance phenomena"

N. Das, Ph.D., Physics, May 2010, Dissertation: "E-field Induced Resistive Switch in Transition Metal Oxide: A Model for Future Nonvolatile Memory Devices"

R. P. Chaudhury, Ph.D., Physics, August 2010, Dissertation: "Effects of High Pressure, Magnetic Fields and Substitutions on Multiferroic Magnetoelectric Systems"

M. Gooch, Ph.D., Physics, December 2010, Dissertation: "A Study of Magnetic Spin Fluctuations and Superconductivity in the Iron Pnictides"

F. Y. Wei, Ph.D., Physics, August 2011, Dissertation: "Thermodynamics investigation of doping and correlation between magnetism and superconductivity in FeAs-based superconductors"

K.-C. Liang, Ph.D., Physics, August 2013, Dissertation: "Effects of magnetic fields and substitutions on magnetoelectric and multiferroic systems"

B. Jawdat, Ph.D., Physics, August 2015, Dissertation: "A Study of Superconductivity in a Series of Strontium Platinum Phosphides"

L. Z. Deng, Ph.D., Physics, August 2015, Dissertation: "Possible Interface Enhanced Superconductivity in Iron Pnictides and Chalcogenides"

K. Shrestha, Ph.D., Physics, December 2015, Dissertation: "Magnetotransport Studies on Topological Insulators"

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Abstract

We have discovered a record Tc up to 49K in the rare-earth doped CaFe2As2 system with RE = La, Ce, Pr, and Nd, highest in the Fe-based 122 superconductor family. Detailed studies have demonstrated the system's non-bulk nature, two superconducting transitions, unusually high magnetic anisotropy, and RE-independent and dopant-concentration-independent nature. The unusually high Tc is attributed to the possible long-sought-after interfacial enhancement mechanism [proposed by Allender-Bray-Bardeen (ABB)] via naturally grown FeAs layers with different defect density, opening up a new avenue to higher Tc (ABB suggested possible room temperature Tc via this mechanism). We have developed a novel technique for detailed magnetic studies of the artificially grown FeSe/STO system, and observed: a Meissner state below ~ 20 K; a mesoscopic state with superconducting patches between ~ 20 and 45 K; and a yet-to-be-determined state between ~ 45 and 80 K with a logarithmic frequency dispersion. We have discovered more than ten new superconductors of new structure

types, with anti-perovskite feature as cuprate, as first superconductor in large compound family, or through novel synthetic routes. These findings will enable our capability and extend our directions to look for more new superconductors with higher Tc. New physics and phenomena were also discovered in both superconducting and non-superconducting compounds such as: superconducting-semiconducting transition in YBCO under high pressure; raising questions regarding topological superconductivity in the $Cu_xBi_2Se_3$; temperature-induced orbital-selective Mott transition (OSMT) in Fe-chalcogenides; field-induced spin-flop transition in the polar compound $LiCrP_2O_7$; and quantum oscillations from topological surface states of metallic $Bi_2(SeTe)_3$. In addition, the studies of multiferroic and magnetoelectric compounds have led to several milestones in the $RAl_3(BO_3)_4$, $Mn_{1-x}Co_xWO_4$, and $LiFeP_2O_7$ materials. Moreover, we have extended our studies to other AFOSR mission-related systems such as super-thermal-conducting materials, thermoelectric materials with higher ZT and better performance, and 2D transition metal dichalcogenides.

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